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PHYSICAL REVIEW B **84**, 121407(R) (2011)

# Spin splitting in graphene studied by means of tilted magnetic-field experiments

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We have measured the spin splitting in single-layer and bilayer graphene by means of tilted magnetic-field experiments. By applying the Lifshitz-Kosevich formula for the spin-induced decrease of the Shubnikov-de Haas amplitudes with increasing tilt angle, we directly determine the product between the carrier cyclotron mass $m^*$ and the effective $g$ factor $g^*$ as a function of the charge-carrier concentration. By using the cyclotron mass for a single-layer and a bilayer graphene, we find an enhanced $g$ factor $g^* = 2.7 \pm 0.2$ for both systems.

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The half-integer quantum Hall effect in single-layer graphene (SLG)[1,2] and the unconventional quantum Hall effect in bilayer graphene (BLG)[3] reveal spin- and valley-degenerate relativistic Landau levels. Due to the extremely large Landau-level splitting,[4,5] completely resolved levels can be observed up to room temperature.[6] However, even at very high perpendicular magnetic fields the Zeeman splitting within one Landau level is negligible smaller compared to the Landau-level splitting and, more importantly, the Landau-level width generally exceeds the spin splitting. Exceptionally, the zeroth Landau level in SLG becomes extremely narrow at magnetic fields $B > 20$ T,[4] which allows an experimental observation of a spin-related gap opening at magnetic fields $B > 20$ T.[7] Another observation of a spin degeneracy lifting with an effective $g$ factor $g^* = 2$ was reported for $\nu = \pm 4$, in SLG for magnetic fields $B > 30$ T, combined with lifting the valley degeneracy at $\nu = \pm 1$.[8]

In this Rapid Communication we determine the spin splitting of broadened Landau levels for SLG and BLG by measuring Shubnikov-de Haas (SdH) oscillations in tilted magnetic fields. This technique allows adjusting the ratio between the spin splitting and the Landau-level splitting by controlling the ratio between a total magnetic field and a component perpendicular to a two-dimensional graphene flake. Using the well-established Lifshitz-Kosevich formula[9,10] we determine the product of the effective $g$ factor and cyclotron mass $m^*g^*$ from the angular dependence of the SdH amplitudes and we find that $g^*$ is enhanced compared to the free-electron value.

We have fabricated field-effect transistors from SLG and BLG by micromechanically exfoliating graphene flakes from graphite. The flakes were deposited on top of a Si/$SiO_2$ wafer, structured into a Hall bar and covered with Au/Ti contacts.[11] Charge carriers are introduced by applying a gate voltage on the conducting Si substrate.

We present a detailed analysis on the spin splitting in a SLG sample made from Kish graphite with a mobility $\mu = 0.8$ V m$^{-2}$ s$^{-1}$ and a BLG sample originating from natural graphite with a mobility $\mu = 0.3$ V m$^{-2}$ s$^{-1}$. Two other devices, one SLG and one BLG sample, showed qualitatively similar results.

To determine the spin splitting we have measured the longitudinal resistances $R_{xx}$ as a function of charge-carrier concentration $n$ at a constant perpendicular magnetic field. We adjusted the total magnetic field $B_{\text{tot}}$ for each tilt angle such that the normal component $B_n$ is the same (see the inset to Fig. 1). The value of $B_n$ was verified by measuring the Hall resistance of the devices in the nonquantized regime.

In Fig. 1 we show the experimental $R_{xx}(n)$ dependencies for SLG at $B_n = 6$ T (a) and for BLG at $B_n = 8$ T (b). $R_{xx}$ shows Shubnikov-de Haas oscillations with maxima whenever the Fermi energy is situated in the middle of a spin- and valley-degenerated Landau level $E_N$, $N = 0,1,2,\ldots$ being the Landau-level index. For the higher Landau levels ($N \geqslant 2$) the longitudinal resistances do not exhibit zero minima, indicating that the level broadening is comparable to the cyclotron energy at these perpendicular magnetic fields.

When increasing $B_{\text{tot}}$ at a constant $B_n$, the oscillation amplitudes for both BLG and SLG are reduced. From this reduction we determined the spin splitting. We use the Lifshitz-Kosevich formula for systems with a general dispersion and we specifically include spin splitting[9,10] with an effective $g$ factor $g^*$ (Refs. 12 and 13) and tilted magnetic fields.[14] The oscillatory contribution to the longitudinal resistance can be described as[2]

$$\tilde{R}_{xx} = A \cos\left(\frac{\hbar}{eB_n}\, S(E)|_{E=E_F} + \pi + \varphi_B\right), \qquad (1)$$

where $S(E)|_{E=E_F}$ is an extremal cross section of the Landau orbits in the $k$ space, $A$ is the oscillation amplitude, and $\varphi_B$ is the Berry phase, $\varphi_B = \pi$ for SLG,[1,2] and $\varphi_B = 2\pi$ for BLG.[3] The amplitude $A$ contains a monotonic $n$-dependent part, a temperature dependence, a $B_n$-dependent contribution, and a damping factor due to spin splitting depending on the total field $B_{\text{tot}}$. At a constant temperature and perpendicular magnetic field this $B_{\text{tot}}$ dependence of the SdH amplitude $A$ for charge carriers with cyclotron mass $m^*$ and effective $g$ factor $g^*$ is given by[12,14]

$$A = A_0(N) \cos\left(\frac{\pi}{2}\,\frac{g^*m^*}{m_e}\,\frac{B_{\text{tot}}}{B_n}\right), \qquad (2)$$

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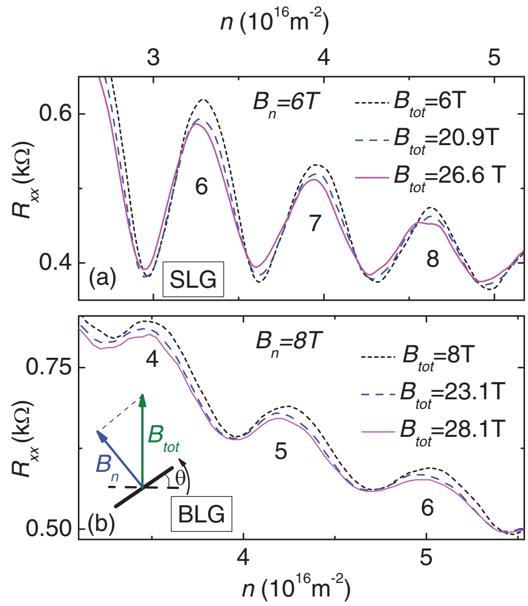


FIG. 1. (Color online) Shubnikov-de Haas oscillations in SLG (a) at $T = 1.3$ K and in BLG(b) at $T = 0.4$ K as a function of the carrier concentration for different total fields $B_{\rm tot}$ or tilt angles $\theta$, respectively. When varying $\theta$, the total field $B_{\rm tot}$ is adjusted such that the perpendicular field $B_n$ remains constant, i.e., $B_{\rm tot} = B_n/\cos\theta$. The oscillation maxima are marked with the corresponding Landau-level numbers $N$. The inset schematically shows this tilting configuration.

with cyclotron mass[1]

$$m^* = \frac{\hbar^2}{2\pi}\frac{dS(E)}{dE}\bigg|_{E=E_F} \tag{3}$$

and $A_0(N)$ is constant for a given $N$.

For the spherical Fermi surface in SLG and BLG with a Fermi wave vector $k_F = \sqrt{\pi n}$, the extremal cross section of the Landau orbits is $S(E)|_{E=E_F} = \pi k_F^2 = n\pi^2$, and Eq. (1) yields the concentration-dependent resistance oscillations as we observe them in our experiments:

$$\tilde{R}_{xx} = A\cos\left(\frac{\hbar\pi^2}{eB_n}n + \pi + \varphi_B\right) = A\cos\left(\frac{\pi}{2}\nu + \pi + \varphi_B\right), \tag{4}$$

where $\nu = (hn)/(eB_n)$ is the filling factor. As expected, the oscillation period $(2eB_n)/(\hbar\pi)$ is independent on the band structure of the two-dimensional material and only depends on the filling factor.

To accurately determine the experimental oscillation amplitudes we have fitted our experimental data $R_{xx}(n)$ to Eq. (4) in two steps. First, we determined the oscillation period and a smooth background using all oscillations measured for a wide range of carrier concentrations. Second, we fitted the oscillation amplitudes $A$ for each individual oscillation using the above determined period and background as fixed parameters. In Fig. 2 we show the final results of this fitting procedure for the SdH amplitude as a function of the total magnetic field for different Landau levels $N$. For clarity, all amplitudes are normalized to $A_0$.

The experimentally observed reduction of the SdH amplitudes can be qualitatively visualized in a simple density of states (DOS) picture of a Landau level as depicted in Fig. 3(a). In a purely perpendicular magnetic field the Landau-level width exceeds the spin splitting and the DOS of the spin-down state [orange, horizontally dashed in Fig. 3(a)] overlaps with the one of the spin-up states (red, vertically dashed) to one broad Landau level. When increasing $B_{\rm tot}$ by leaving $B_n$ constant, these two states move apart, yielding an additional broadening of the Landau level with a reduced DOS in the center [green, solid areas in Fig. 3(a)]. Eventually, when the spin splitting exceeds the level width, a minimum between two distinct levels starts to develop in the DOS. This scenario is indeed observed experimentally in SLG [Fig. 3(b)]. The SdH maxima corresponding to the $N = 9$ and $N = 10$ Landau levels at $B_{\rm tot} = B_n = 5$ T do not show any splitting. Increasing

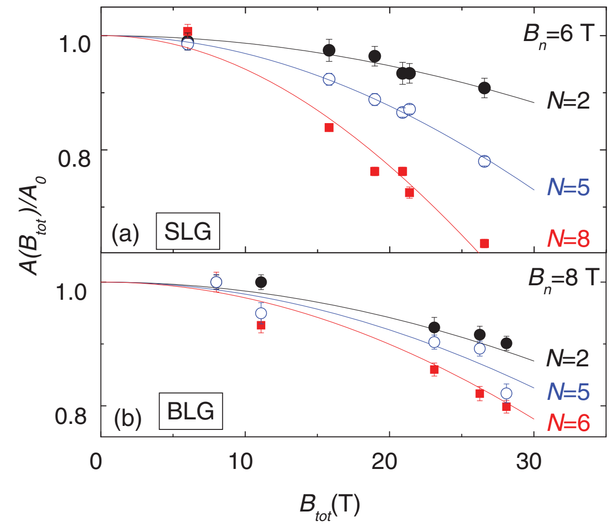


FIG. 2. (Color online) Normalized oscillation amplitudes as a function of total field $B_{\rm tot}$ at a constant perpendicular field $B_n$ in SLG (a) and BLG (b). Error bars represent standard least-squares-fitting errors in the determination of $A$. Solid lines are fits to Eq. (2) with $m^*g^*$ as a fit parameter.

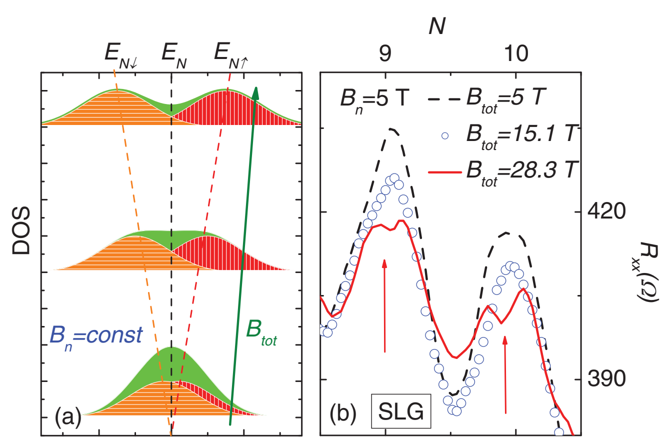


FIG. 3. (Color online) Schematic representation of the density of states for a Landau level with an increasing total magnetic field $B_{\rm tot}$ (from the bottom to the top) at a constant perpendicular component $B_n$ (a). (b) shows this scenario as measured experimentally for the $N = 9,10$ maximum in SLG at a constant perpendicular magnetic field $B_n = 5$ T.

the total field at a constant perpendicular component leads to a reduction of the oscillation amplitude and eventually to the appearance of spin-resolved peaks at the highest field of 28 T. However, this splitting is not yet enough to determine the energy difference by, e.g., activation measurements.

A quantitative analysis of this decrease of the SdH amplitudes with increasing total magnetic field is done by fitting the data to Eq. (2) with $m^*g^*$ as a fitting parameter (solid lines in Fig. 2). The values for $m^*g^*$ obtained are plotted as a function of the charge-carrier concentration in Fig. 4 for SLG (a) and BLG (b).

For both SLG and BLG the product $m^*g^*$ increases with concentration, which can be mainly attributed to the concentration-dependent cyclotron mass $m^*$ of particles with a linear[1] and hyperbolic dispersion[15] as predicted by Eq. (3).

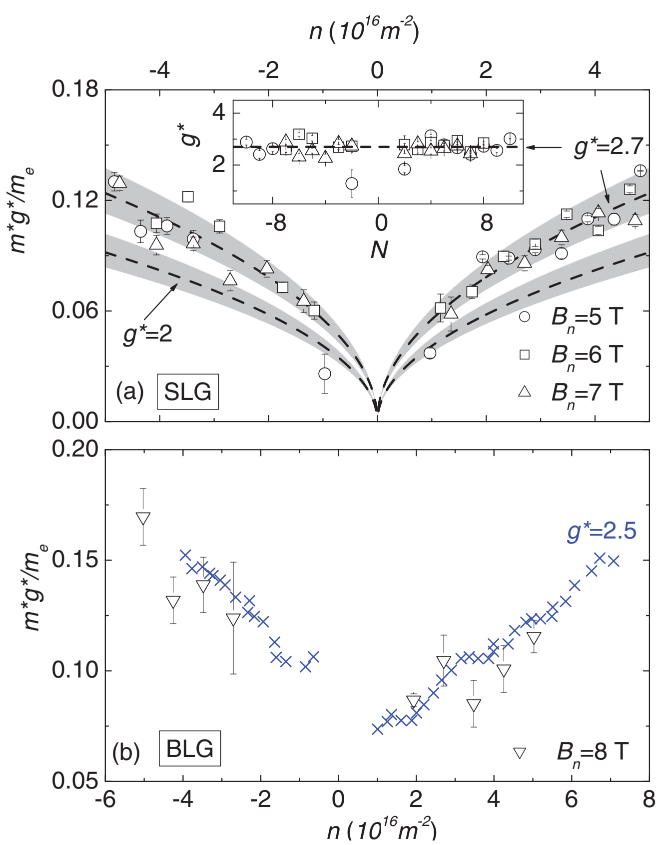


FIG. 4. (Color online) Experimentally deduced $m^*g^*$ (open symbols), normalized to the free-electron mass $m_e$, as a function of charge-carrier concentration for SLG (a) and BLG (b). The individual data points were extracted from the total-field dependence of the SdH amplitudes corresponding to different Landau levels $N = 2, \ldots, 10$ and represent measurements for a constant magnetic field $B_n = 5$, 6, and 7 T for SLG and $B_n = 8$ T for BLG. The error bars represent the standard least-squares-fitting errors, taking into account the error bars of $A$ (Fig. 2). The dashed lines in (a) represent the calculated behavior of $m^*g^*$ for different values of $g^*$, taking into account a 10% experimental uncertainty (shadowed areas). The crosses in (b) compare our data to the experimental cyclotron mass for BLG (Ref. 17) multiplied by $g^* = 2.5$. The inset shows the effective $g$ factor, extracted from the product $m^*g^*$ in the main panel and the known cyclotron mass $m^*$ in SLG, as a function of Landau-level index $N$.

The dashed lines in Fig. 4(a) show the calculated dependence of $m^*g^*$ for $g^* = 2$ and $g^* = 2.7$ using $m^*(n) = (\hbar/c)\sqrt{\pi n}$.[1] The shadowed areas represent a 10% uncertainty of this calculation, mainly due to the experimental errors and some uncertainty in the Fermi velocity.[16]

For SLG [Fig. 4(a)], the increase of $m^*g^*$ with $n$ is symmetric for electrons and holes (i.e., negative and positive $n$ in the figure). A best fit using $m^*(n)$ for SLG yields $g^* = 2.7 \pm 0.2$ (the error is the standard deviation). This finding is shown directly in the inset of Fig. 4(a), where we plot the value of $g^*$ determined in the middle of each Landau level $N$ for different perpendicular fields $B_n$. Within an experimental error $g^*$ does not show any dependence on $N$ or $B_n$.

For BLG [Fig. 4(b)] the experimental situation is more complex as the observed increase of $m^*g^*$ with $n$ is not symmetric for holes and electrons. Such a behavior is caused by an asymmetry of $m^*$ resulting from an asymmetric band structure of biased BLG, which was already observed experimentally in transport experiments,[17] cyclotron resonance,[18] and activation-gap measurements.[5] Applying the experimental cyclotron mass from Ref. 17 (depicted as crosses in Fig. 4) allows us to estimate $g^*$ to be $\sim$2.5 for both electrons and holes which is, within experimental accuracy, reasonably consistent with the $g$-factor enhancement observed in SLG.

The observed enhancement of the effective spin splitting compared to its free-electron value can be explained by an electron-electron interaction[19] yielding an interaction-enhanced splitting between two spin levels within one Landau level:[20,21]

$$g^*\mu_B B_{\text{tot}} = g\mu_B B_{\text{tot}} + E^0_{\text{ex}}(n_\downarrow - n_\uparrow). \tag{5}$$

Here $g = 2$ is a free-electron $g$ factor, $E^0_{\text{ex}}$ is an exchange parameter, and $n_\uparrow$ and $n_\downarrow$ are the relative occupations of the two spin states of a given Landau level.

For Gaussian-shaped Landau levels with broadening $\Gamma > g^*\mu_B B_{\text{tot}}$, i.e., where the spin splitting is not yet resolved, this relative occupation difference can be approximated by using the Taylor expansion of the Gauss error function $\text{erf}(g^*\mu_B B_{\text{tot}}/\Gamma)$:

$$n_\downarrow - n_\uparrow \approx \sqrt{\frac{1}{2\pi}}\,\frac{g^*\mu_B B_{\text{tot}}}{\Gamma}, \tag{6}$$

and Eq. (5) yields

$$\frac{g^*}{g} = \left(1 - \sqrt{\frac{1}{2\pi}}\frac{E^0_{\text{ex}}}{\Gamma}\right)^{-1}. \tag{7}$$

$E^0_{\text{ex}}$ is of the order of the Coulomb interaction $E^0_{\text{ex}} \propto \sqrt{B_n}$,[21] and $\Gamma \propto \sqrt{B_n}$.[22] Therefore, the ratio $E^0_{\text{ex}}/\Gamma$ remains constant and the $g$-factor enhancement is indeed predicted to be constant, as we observe experimentally. Using the experimentally found $g^* = 2.7$ in Eq. (7) yields $E^0_{\text{ex}} = 130$ K at 10 T when assuming $\Gamma = 200$ K.[4,5] For a completely spin-polarized system, i.e., $n_\downarrow - n_\uparrow = 1$, one might then speculate that the exchange enhancement in Eq. (5) would be an order of magnitude larger than a single-particle Zeeman energy at this particular field.

Finally, we note that the experimentally found enhanced values of $g^*$ in graphene are close to those observed in transport experiments in graphite.[23] This may suggest that an exchange-induced enhancement of $g^*$ is quite common for graphitic materials. In contrast, no interaction-induced $g$-factor enhancement is observed using electron-spin resonance in graphene[24] and graphite[25] since these measurements are not sensitive to many-body corrections.[26] Interestingly, measuring the Zeeman splitting of single-electron states in quantum dots, where no exchange enhancement of the $g$ factor is expected, also yields $g \approx 2$,[27] albeit with a considerable experimental uncertainty.

To conclude, we have experimentally measured and analyzed spin splitting in SLG and BLG. We have shown that the product between the cyclotron mass $m^*$ and the effective $g$ factor $g^*$ increases with charge-carrier concentration, as expected for a linear dispersion in SLG and a hyperbolic dispersion in BLG. Using the known concentration dependence of $m^*$, we found that $g^*$ in graphene is enhanced compared to the free-electron value, and we attribute this to electron-electron interaction effects.

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